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REVIEW ARTICLE

Valorization of fruits and vegetable wastes and by-products to produce natural pigments

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ABSTRACT

Synthetic pigments from petrochemicals have been extensively used in a wide range of food products. However, these pigments have adverse effects on human health that has rendered it obligatory to the scientific community in order to explore for much safer, natural, and eco-friendly pigments. In this regard, exploiting the potential of agri-food wastes presumes importance, extracted mainly by employing green processing and extraction technologies. Of late, pigments market size is growing rapidly owing to their extensive uses. Hence, there is a need for sustainable production of pigments from renewable bioresources. Valorization of vegetal wastes (fruits and vegetables) and their by-products (e.g. peels, seeds or pomace) can meet the demands of natural pigment production at the industrial levels for potential food, pharmaceuticals, and cosmeceuticals applications. These wastes/by-products are a rich source of natural pigments such as: anthocyanins, betalains, carotenoids, and chlorophylls. It is envisaged that these natural pigments can contribute significantly to the development of functional foods as well as impart rich biotherapeutic potential. With a sustainability approach, we have critically reviewed vital research information and developments made on natural pigments from vegetal wastes, greener extraction and processing technologies, encapsulation techniques and potential bioactivities. Designed with an eco-friendly approach, it is expected that this review will benefit not only the concerned industries but also be of use to health-conscious consumers.

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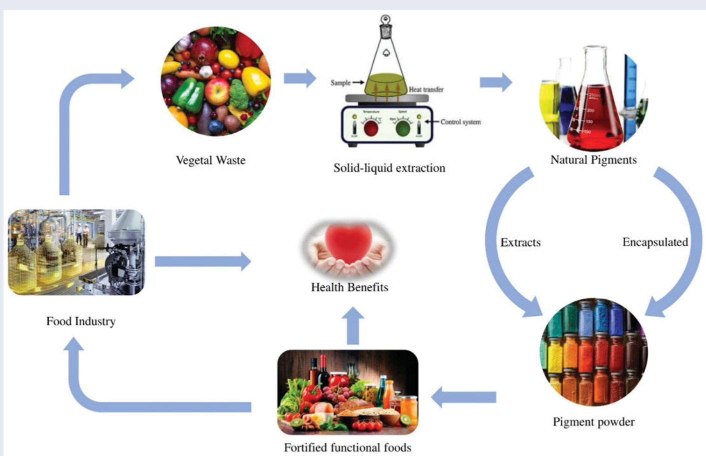
KEYWORDS
Natural pigments; vegetal waste; valorization; green extraction technologies; encapsulation; bioactivity

GRAPHICAL ABSTRACT

Schematic representation of vegetal waste utilization for the exploitation of the pigments and their functional properties.



HIGHLIGHTS

- Valorization of vegetal wastes and by-products to produce natural pigments
- Recent developments in green-extraction techniques for the isolation of natural pigments
- Encapsulation of pigments from vegetal wastes
- Natural pigments impart rich health benefits.

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Introduction

Historically, natural pigments or colorants used in food applications were obtained from renewable resources such as from plants or from microbes (e.g. bacteria, algae, fungi, and yeasts) and insects. Extracts of: saffron, paprika, turmeric, and various flowers are some examples from which natural pigments were traditionally derived [1]. With wide profound applications, synthetic pigments gained popularity as one of the main food coloring (pigments) compounds. These synthetic compounds have been related with high stability, low production costs, and high tinctorial strength as well as ease of application in the food system. Edible pigments are ubiquitously present in plant origin foods, especially in colored fruits and vegetables (Table 1). Nature provides a wide array of colors to fruits and vegetables, and thus is a pigment enriched food resource. These pigments occur mainly as secondary plant metabolites in fruits and vegetables and are usually coined as natural pigments. Nowadays, an increasing trend has been witnessed wherein consumers are more concerned about the diet pattern they consume and their health impact.

Owing to consumers demand, considerable innovative research has been undertaken to explore for natural and safe food ingredients such as natural pigments with potential health benefits (Table 1) [2–53]. This has rendered it a necessity to find alternatives for chemical-based pigments [54]. Researchers are continually looking for natural food coloring alternatives to meet the market challenges and demands, and those which can fulfill and meet regulatory restrictions for food and biotherapeutics applications.

The food processing industry produces enormous amount of waste and by-products, and these form the second-largest generator of wastes after household sewage wastes [55]. Processing waste generation is continuously increasing with increased industrialization and urbanization. Fruit and vegetable wastes and/or by-products obtained from the processing industries mainly contains the skin portion or the peel, seeds, and pomace. These are excellent resources for ingredients such as protein, peptides, polysaccharides, dietary fibers, and others along with bioactive-functional ingredients such as polyphenols, antioxidants and antimicrobial compounds including natural pigments [56]. Among all of these, pigments are considered to be natural, safe, and possess potential antioxidant activities, as well as can be a potential coloring material source in food applications [57]. These natural pigments, with coloring and pharmacological properties, have wide applications in the food industry and can be utilized in product development (functional and nutraceutical foods) because of their health-related beneficial effects [58]. On the other hand, due to the shortage of pilot testing of the innovative technologies, the exploitation of the vegetal wastes for pigment extraction has been highly limited [55]. Various innovative techniques, such as pulsed-electric field, pulse-light, high pressure processing, ionizing radiation have been explored for the extraction of new potential food colorants from vegetal wastes [59]. The wastes contain significant amounts of phyto-pigments which can help in overcoming cardiovascular, cerebrovascular, and certain types of cancers [60]. These natural pigments can be isolated from wastes and by-products and can possess potential bioactivities such as antimicrobial, antioxidant, antiproliferative, anti-inflammatory properties, etc. [60]. Due to significant nutritional and beneficial health properties, as well as phyto-pigment-antioxidant nature, these plants secondary metabolites can be considered as 'functional food ingredients'.

In this review, the focus is on the major types of natural pigments in vegetal wastes, effective utilization of vegetal wastes for extraction of natural pigments, and encapsulation technologies to enhance the stability of natural pigments, as well as their applications during the development of novel functional foods. From a biotechnological viewpoint, recent developments on the utilization of vegetal wastes for the exploitation of natural pigments as bioactives in a sustainable manner is also highlighted.

Designed with an eco-friendly environmental concern based approach and keeping in mind circular economy concepts, it is expected that this review will benefit not only the dependent industries but also be of use to health-conscious consumers.

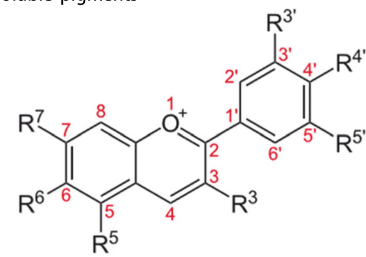
Natural pigments – types and sources

Pigments are classified as either natural or synthetic, water and/or fat-soluble, organic or inorganic types. Further, they are also classified based on their structural affinities, solubility and natural occurrence. The classification of natural pigments primarily extracted from vegetal wastes are specified in the Figure 1. Most often, these are mainly divided into 04 major groups: anthocyanins, betalains, carotenoids and chlorophyll (see Table 1) [61].

Anthocyanins

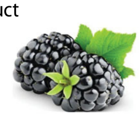
Anthocyanins are natural, water-soluble, nontoxic, vacuolar and the largest group of polyphenolic pigments.

Table 1. Natural pigments extracted from vegetal wastes.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
			Water soluble pigments		
Anthocyanins					
Apple (*Malus domestica*) peel 	Solvent extraction	80% acetone or ethanol	169.7g cyanidin 3-glucoside equivalents/100 g dried peels	Antioxidants, Free radicals scavenger, Anti-inflammatory, Anti-viral, Anti-cancer properties	[2]
Banana (*Musa* sp.) peel 	Solvent extraction	Methanol, ethanol, acetone, water, acetone:water, methanol:water or ethanol:water	434 µg cyanidin 3-glucoside equivalents/100 g d.w.	Antioxidant activity Development of drugs and use in functional foods. Free-radical scavenging properties	[3–4]
Eggplant (*Solanum melongena*) peel 	Solvent extraction	70% methanol, 70% ethanol and 70% acetone	Methanol- 82.83, Ethanol- 62.92, Acetone-51.56 mg DGE/100 g DP	Color (pigments) and antioxidant properties	[5]
Blackberry (*Rubus* sp.) residue 	Solvent extraction	Acidified ethanol with citric acid	4.31 mgCy3GlE/g de	Therapeutic potential, antioxidant, anti-inflammatory, antimicrobial, gastroprotective, and cardioprotective properties	[6–7]
Grape (*Vitis vinifera*) skin 	Solvent extraction	0.01% HCl acidified 70% aqueous acetone followed by chloroform and evaporation at 30 °C	987.9 to 382.1 mg Mv^3G/100 g of skin (of four varieties)	Color/pigments Antioxidant, cardioprotective, anticancer, anti-inflammation, antiaging and antimicrobial activities, which are making greater potential for grape in the field of food and pharmaceutical applications	[8]

(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Blackcurrant (*Ribes nigrum*) waste 	Water extraction	Acidified water extraction followed by a solid-phase extraction (SPE)	2% w/w of d.w.	Antioxidant activity	[9]
Blackberry (*Rubus* sps.) by-product 	Water extraction	Water	Extract- 718.47, Concentrate- 389.09 mg cyd 3-glu/ 100 g db	Antioxidant and antimicrobial activities	[10]
Red grape (*Vitis vinifera*) pomace 	Water extraction	Hot water extraction and expeller	Dried samples- 41%, Fresh samples- 68% of monomeric anthocyanins	Anti-inflammatory, anti-angiogenic and redox properties	[11]
Coffee (*Coffea* sps.: *C. Arabica* and *C. robusta*) exocarp 	Ethanol extraction	60% ethanol	0.145 mg cyanidin 3-glucoside/g	Anti-oxidant, anti-carcinogenic, anti-inflammatory agents and anti-hypoglycemic activities	[12]
Eggplant (*Solanum melongena*) peel 	Water extraction	Water (80 °C for 40 min)	–	Antioxidant, anti-carcinogenic, anti-inflammatory, anti-viral, and anti-diabetic activities	[13]
Sour cherry (*Prunus cerasus*) pomace 	Ethanol-water extraction	50% ethanol	WE10- 67.92, SE10- 64.33, WE15- 58.75 SE15-35.91%	Antioxidant, antimicrobial activities, natural colorant (pigments), health promoting dietary fiber and can be used as food thickener.	[14]
Grape (*Vitis vinifera*) skin extracts 	Supercritical CO_2 extraction	100–130 bar; 30–40 °C; pH- 2–4; ethyl alcohol – 25–30%; CO2 flow rate 25–50 ml/min	80–85% recovery	Antioxidant, cardioprotective, anticancer, anti-inflammation, antiaging and antimicrobial properties.	[15]

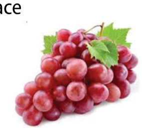
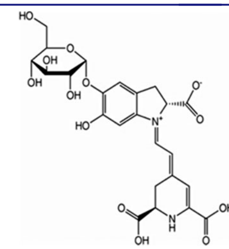
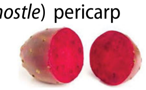
(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Grape (*Vitis vinifera*) peel 	Supercritical fluid extraction (SFE)	45–46 °C, 160–165 kg cm^{-2} pressure and 6–7% ethanol	1.176 mg/mL	Radical scavenging activity	[16]
Grape (*Vitis vinifera*) by-products 	Combined extractions	Water 70 °C; Ultrasonics (35 KHz); HHP (600 MPa); PEF (3 kV cm^{-1})	7.93, 7.76, 11.21 and 14.05 mg Cy-3-glu eq. g^{-1} DM	Antibacterial, antiviral, antioxidant, anti-inflammatory, anti-cancerogenic properties. Prevent cardiovascular diseases	[17]
Jaboticaba (*Plinia cauliflora*) peel 	Ultra-sound-assisted extraction	Hydroalcoholic mixture (50 v/v) + ultrasonic bath (40 kHz and 150 W)	3.4 mg/g raw material	High antioxidant activity	[18]
Jaboticaba (*Plinia cauliflora*) pomace 	Ultra-sound-assisted extraction	Water, 531 W $cm-2$, for 15 min at 20 °C	510.35 mg of cyanidin-3-glicosideo 100 $g-^{1}$	Multiple health benefits, including the prevention and/or mitigation of oxidation, inflammation, atherosclerosis risk factors, cancer, and conditions involved with the metabolic syndrome.	[19]
Jaboticaba (*Plinia cauliflora*) peel 	Pressurized Liquid Extraction (PLE) + supercritical CO_2	5 MPa, 80 °C, 9 min in presence of carbon dioxide, ethanol	–	Strong antioxidant, anti-inflammatory, anti-diabetic, and anti-obesity properties. Useful in the treatment of chronic obstructive pulmonary disease	[20]
Acerola (*Malpighia emarginata*) residue 	Ultra-sound-assisted extraction UAE	Acidified ethanol with ultrasound mixer	Anthocyanin: 2.00 to 11.16 mg TA/100 g	High antioxidant capacity. Bio-functional properties like skin whitening effect, anti-aging and multidrug resistant reversal activity.	[21]
Purple potato (*Solanum tuberosum*) peel 	Ultra-sound-assisted extraction UAE	Ultrasounds for 20 min, 30 °C with methanol–acetone–water (7:7:6, v/v/v)	6.84 mg/100 g	Antioxidants, anti-inflammatory, anti-mutation, and anti-tumor properties.	[22]

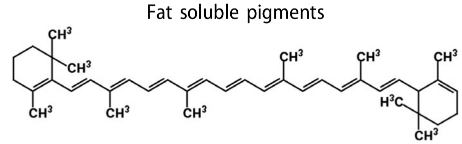
(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Sweet cherry (*Prunus avium*) skin 	Ultra-sound-assisted extraction (UAE)	70% ethanol, 40 kHz, 100 W, at 40 °C	12.2 mg/g	Strong antioxidant capacity, neuroprotective effects, cancer preventive properties, pain relief from inflammation and arthritis	[23]
Red grape (*Vitis vinifera*) pomace 	Combined extractions	Water and Ethanol, separately UAE- water and water: ethanol (1:1), 25 kHz frequency, 300 W at 20 °C for 60 min. MAE- water and water: ethanol (1:1), 50 °C, power at 200 W, 60 min	UAE −34188 ppm	Anti-inflammatory, anti-angiogenic and redox properties	[24]
Betalains					
Red Pitaya *(Hylocereus polyrhizus*) peel 	Solvent extraction	80% acetone	13.8 mg as betanin equivalents per 100 g	Alternatives to synthetic colorants as they possess strong coloring potential, Excellent health-contributing properties.	[25–26]
Prickly pear (*Opuntia joconostle*) pericarp 	Solvent extraction	80% methanol containing 0.1% HCl	4.56 mg betanin/100 g	Antioxidant activity	[27–28]
Ulluco (*Ullucus tuberosus*) peel 	Solvent extraction	methanol: water (60:40) for 24 h at 10 °C	100 µg/g	Scavenging of free radicals/reactive oxygen species, Inhibition of lipid peroxidation and LDL oxidation, prevention of DNA damage. Induction of antioxidant, anti-proliferative and anti-microbial activities.	[29–30]

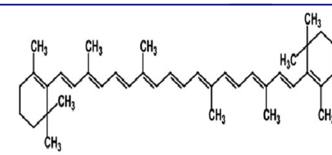
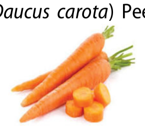
(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Beetroot (*Beta vulgaris*) peel and pomace 	Ultra-sound-assisted extraction (UAE)	50% aqueous ethano*l* + 0.5% acetic acid at ultrasonic bath (50/60 Hz, 125 W, 22 °C)	Peel- 3.8–7.6 mg/g Pomace- 37.22 mg/100 g	Antioxidant and antiproliferative activities, free radical scavengers that prevent active oxygen-induced and free radical-mediated oxidation of biological molecules.	[31]
Pitaya or dragon fruit (*Hylocereus undatus*) peel 	Ultra-sound-assisted extraction (UAE)	80% acetone in an ultrasound/15 min	101.04 mg/100g	Antioxidant capacity and anti-bacterial activity.	[32]
Pitaya (*Hylocereus undatus*) peel 	Microwave-assisted extraction (MAE)	Ethanol and water (40:60, v/v) microwave oven	–	Antioxidant capacity, antibacterial activity.	[33]
Carotenoids			Fat soluble pigments 		
Tomato (*Solanum lycopersicum*) peel 	Solvent extraction	Acetone and hexane (50:50)	–	Antioxidant, Anti-mutagenic, anti-proliferative, anti-inflammatory and anti-atherogenic activities. Carotenoids helps to modulate immune response, stimulate intercellular signaling (gap junction) pathways, possess pro-vitamin A activity, regulate cell cycle and apoptosis, and modulate many physiological processes.	[34]
Tomato (*Solanum lycopersicum*) Skin and pomace 	Solvent extraction	Ethyl acetate: hexane	37.5 mg/ kg dry waste	Natural food colorant, Radical scavenging activity, Reducing antioxidant power.	[35–36]
Gac (*Momordica cochinchinensis*) peel 	Solvent extraction	Hexane/acetone/ethanol (50:25:25 v/v/v)	95 to 136 mg/100 g DW	Antioxidant activity, β-carotene a precursor to vitamin A and has been efficiently used to treat the deficiency of vitamin A through the addition of Gac aril in the diet.	[37]

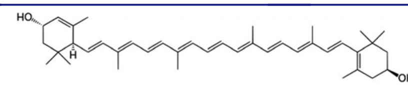
(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Cashew apple (*Anacardium occidentale*) by-products 	Mechanical-press extraction	Helical-type press	6 mg/kg	Rich source of vitamin C, and natural antioxidants. Provides protection against some degenerative diseases and decreases the risk of chronic diseases. Neutralizing free radicals and reduce oxidative damage in the body.	[38]
Pomegranate (*Punica granatum*) peel 	UAE-green solvents	Sunflower and soy oil	0.6134 and 0.6715 mg carotenoids/100 g of dry peels using sunflower oil and soy oil, respectively	Antioxidant, anti-mutagenic, anti-hypertension, anti-inflammatory, anti- -atherosclerotic, osteoarthritis, prostate cancer, heart disease, and anti HIV1 activities.	[39]
Acerola (*Malpighia emarginata*) residue 	Ultra-sound-assisted extraction UAE	Acidified ethanol with ultrasound mixer	Carotenoids-0.76 to 1.58 µg β-CE/g	High antioxidant capacity and Bio-functional properties like skin whitening effect, anti-aging and multidrug resistant reversal activity.	[21]
Beta carotene					
Orange (*Citrus X sinensis*), Pomegranate (*Punica granatum*) and Mango (*Mangifera indica*) Peels 	Solvent extraction	Ethanol: hexane (4:3)	MP-3.16, OP-4.99, PP-2.47 mg/100g	Antioxidant activity, Lowers the risk of developing cancer and heart disease. Precursor of vitamin A (Retinol). Ameliorates oxidative stress induced by reactive oxygen species in living animal systems.	[40–41]
Carrot (*Daucus carota*) Peel 	Solvent extraction	Ethanol + 2 N potassium hydroxide	Before drying- 20.45 mg/100 g dry weight, After drying and blanching- 11.11 mg/100 g dry weight	Oral sun protectant for the prevention of sunburn. Anti-cancer agents, potential treatment of leukemia.	[42]
Tomato (*Solanum lycopersicum*) peel 	Solvent extraction	Sample to solvent ratio (1:5 w/v) with methanol, ethanol, diethyl ether, acetone, chloroform, and hexane	6.87 mg/g of extract	Antioxidant activity	[43–44]

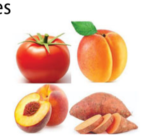
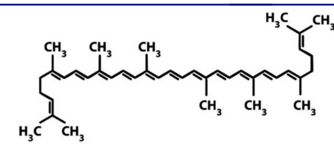
(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Tomato (*Solanum lycopersicum*) by-products 	Supercritical CO_2 extraction	a)50–80 °C, 300–500 bar and 3–6 g CO_2/min flow rates for 105 min b) 2 h, CO_2 flow rate- 4 kg/h at 55 °C and 300 bar, 5% ethanol	28.38% to 58.8% for β-carotene 53.93%	Antioxidant and singlet oxygen quencher	[45]
Pumpkin (*Cucurbita* sps.) seeds 	Supercritical CO_2 extraction	30 MPa pressure, temperature of 47.75 °C and 67% of seed	18.50 mg/100 g sample	Health-promoting benefits Cholesterol lowering effects	[46]
Sweet potato (*Ipomoea batatas*), tomato (*Solanum lycopersicum*), apricot (*Prunus armeniaca*), pumpkin (*Cucurbita pepo*), peach (*Prunus persica*) peels and pepper (*Piper nigrum*) wastes 	Supercritical fluid extraction	$T = 59$ °C, Pressure = 350 bar, $EtOH = 15.5\%$, CO2 flow rate = 15 g/min, run time: 30 min	91–99.8 % recovery for all the samples as compared with solvent extraction	Antioxidant activity	[47]
Lutein					
Tomato (*Solanum lycopersicum*) peel 	Solvent extraction	Sample to solvent ratio (1:5 w/v) with methanol, ethanol, diethyl ether, acetone, chloroform, and hexane	1.08 mg/g of extract	Antioxidants, Contributes toward maintenance of skin health.	[43–44]
Spinach (*Spinacia oleracea*) by-products 	Ethanol extraction	Acetone as conventional method, and 93% ethanol, for 4.3 h at 43 °C as green extraction method	For Acetone- lutein-10.1 mg/100g, For Ethanol- lutein-70% retention	Health benefits involving age related macular degeneration, cardiovascular and cancer diseases. Anti-inflammatory and anti-oxidant properties	[48–49]
Paprika (*Capsicum annuum*) leaves 	Ethanol extraction	79.63% ethanol	232.60 µg/g	commercial applications in nutraceuticals and pharmaceuticals based products to reduce the risk of age-related macular degeneration.	[50]

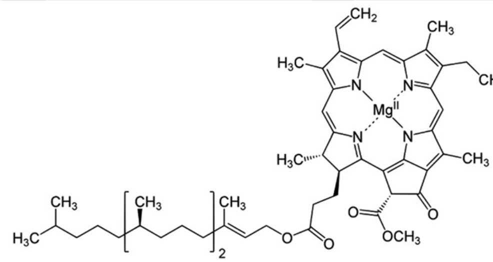
(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Sweet potato (*Ipomoea batatas*), Tomato (*Solanum lycopersicum*), Apricot (*Prunus armeniaca*), pumpkin (*Cucurbita pepo*), Peach (*Prunus persica*) peels and Pepper (*Piper nigrum*) wastes 	Supercritical fluid extraction	$T = 59$ °C, $Pressure = 350$ bar, $EtOH = 15.5\%$, CO2 flow rate = 15 g/min, run time: 30 min	91–99.8 % recovery for all the samples as compared with solvent extraction	Antioxidant activity High in health promoting dietary fiber.	[47]
Lycopene					
Tomato (*Solanum lycopersicum*) by-products 	Supercritical CO_2 extraction	a)50–80 °C, 300–500 bar and 3–6 g CO_2/min flow rates for 105 min b) 2 h, CO_2 flow rate- 4 kg/h at 55 °C and 300 bar, 5% ethanol	32.02% to 60.85% for lycopene and from 28.38% to 58.8% for β-carotene 53.93%	Protection of humans against prostate and other cancers	[45] [51]
Sweet potato (*Ipomoea batatas*), Tomato (*Solanum lycopersicum*), apricot (*Prunus armeniaca*), Pumpkin (*Cucurbita pepo*), Peach (*Prunus persica*) peels and Pepper (*Piper nigrum*) wastes 	Supercritical fluid extraction	$T = 59$ °C, $Pressure = 350$ bar, $EtOH = 15.5\%$, CO2 flow rate = 15 g/min, run time: 30 min	91–99.8 % recovery for all the samples as compared with solvent extraction	Antioxidant activity	[47]
Tomato (*Solanum lycopersicum*) skin 	Ultra-sound-assisted extraction (UAE)	Ethyl acetate (100%)	7.23 g kg^{-1} of tomato peel	Functional ingredient in the formulation of antioxidant rich functional foods Plays a key role in the health protection mechanisms by scavenging free radicals A great source of trace elements, like selenium, copper, manganese and zinc, which are cofactors of antioxidant enzymes Has a potential role in the prevention of chronic diseases	[52]

(*continued*)

Table 1. Continued.

Type of pigment and Raw material	Method of extraction	Processing conditions	Yield	Bioactivity and applications	Reference
Tomato (*Solanum lycopersicum*) peel 	Enzyme-assisted extraction (EAE)	Hexane, ethyl acetate and the mixture hexane: acetone: ethanol, (50:25:25, v/v) with enzymes	440 mg per 100 g of dry peel	Used as a food additive as colorants and Reduces the risk of cardiovascular diseases, atherosclerosis, prostate cancer and cognitive impairment	[53]
Tomato (*Solanum lycopersicum*) peel 	Solvent extraction	Sample to solvent ratio (1:5 w/v) with methanol, ethanol, diethyl ether, acetone, chloroform, and hexane	Cis-lycopene-22.02 mg/g of extract Trans-lycopene-36.49 mg/g of extract	Lycopene supplementation (2.5–10 µM) reduced total cholesterol by decreasing HMG-CoA reductase expression. Lycopene content in blood is known to be inversely proportional to the incidence of heart diseases. Regular consumption is inversely correlated with the risk of inflammatory disorders such as atherosclerosis. Oxidative modulation of low-density lipoprotein (LDL) plays a major role in protection against atherosclerosis and CVD.	[43–44]
Chlorophyll					
Cucumber (*Cucumis sativus*) peel 	Solvent extraction	Sample to solvent ratio (1:5 w/v) with methanol, ethanol, diethyl ether, acetone, chloroform, and hexane	3.46 mg/g of extract	Antioxidant and antimicrobial activities	[43–44]
Watermelon (*Citrullus lanatus*) Peel 	Solvent extraction	Sample to solvent ratio (1:5 w/v) with methanol, ethanol, diethyl ether, acetone, chloroform, and hexane	5.28 mg/g of extract	Antioxidant potential Imparts green color to the melon and can be natural source of pigments to be used in food and pharmaceutical industries	[43–44]
Spinach (*Spinacia oleracea*) by-products 	Ethanol extraction	Acetone as conventional method, and 93% ethanol, for 4.3 h at 43 °C as green extraction method	For Acetone- chlorophyll-12.8 mg/100g For Ethanol-chlorophyll-96%	Chlorophyll: Anti-oxidant, anti- inflammatory and anti-mutagenic properties. Prevents colorectal cancer.	[48–49]

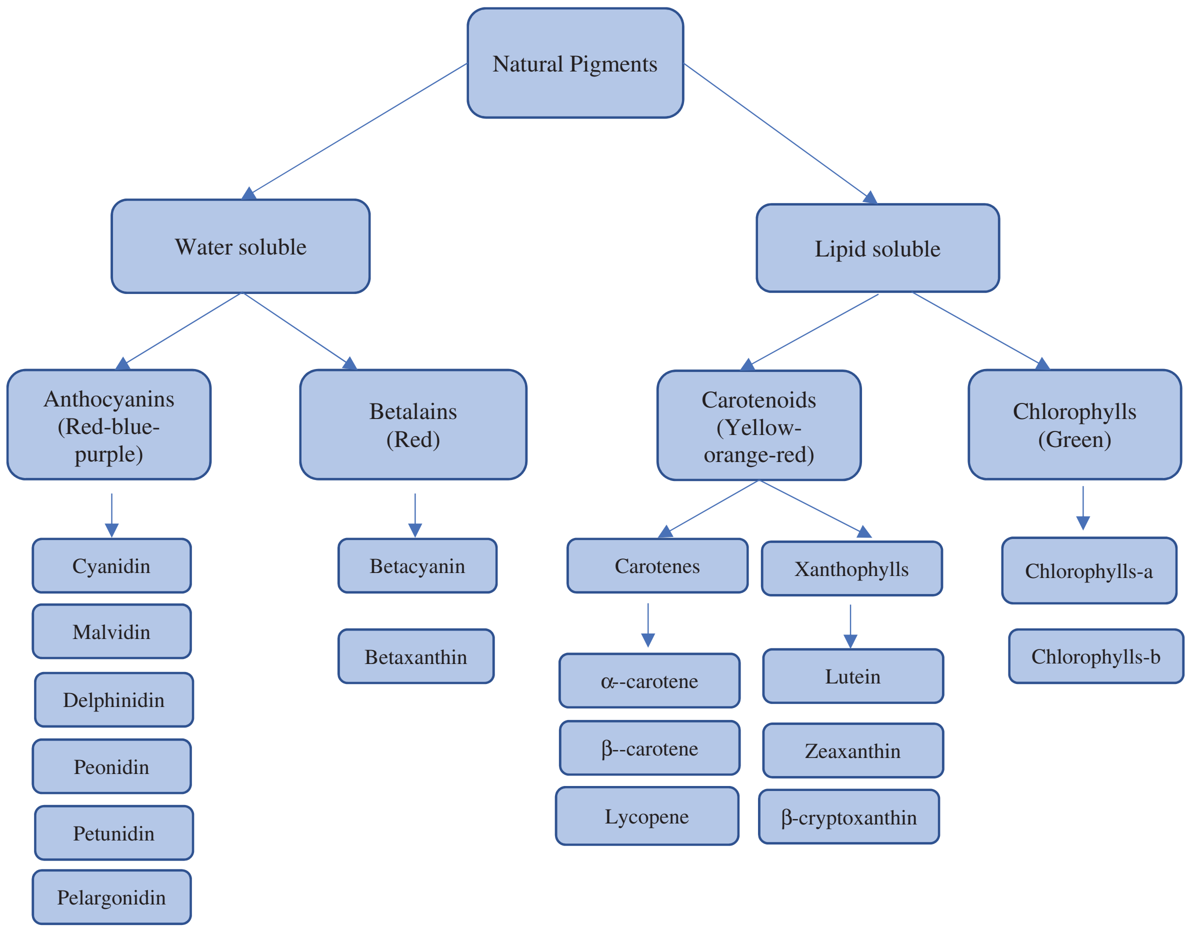


Figure 1. Classification of natural pigments extracted from vegetal wastes.

These are, known to be nearly 700 distinctive structures of anthocyanins [62]. Among vegetables- red cabbage, black carrot, red radish, purple sweet potatos and amongst fruits- blackcurrants, cherries, and berries (e.g. strawberries, elderberries, blackberries, chokeberries, black raspberries, blueberries, black goji berries, concord grapes etc.) have been identified as rich sources of anthocyanins. Anthocyanins belong to the class of flavonoids with the major plant-based anthocyanins being: cyanidin, pelargonidin, delphinidin, peonidin, petunidin and malvidin. The color difference in anthocyanins is due to the structural differences in hydroxyl groups, their number and the position of sugar moieties [9].

Wastes and by-products from food processing industries, like wine or the juice industry, are considered to be additional enriched sources for anthocyanin pigments, which can be exploited as natural colorants for various food applications [55]. Juice processing industries inevitably produce waste and by-products. Blackberry residues are reported to be important sources of natural colorants and nutraceuticals due to the presence of high anthocyanin contents (4.31 mg Cy3GlE/g) [6]. Apple peel can also be a good source of this valuable pigment. The anthocyanin content of Rome beauty apple peels was reported to be 169.7 mg cyanindin 3-glucoside equivalent/100g of dried peel powder [2]. Grape pomace is a well-known source of polyphenolic compounds including: anthocyanins, proanthocyanidins, trans-resveratrol and quercetin [63]. Barros et al. [18], extracted anthocyanins (3.4 mg/g raw material) from freeze-dried jaboticaba peel powder by using an hydroalcoholic mixture (50 v/v), of three different acids (formic acid- 88%, acetic acid- 99.7%, and ortho-phosphoric acid- 85%) at three pH ranges (1.0, 2.0 and 3.0), followed by an ultrasonic bath (frequency- 40 kHz and power- 150 W).

Betalains

Betalains are next to anthocyanins in terms of their naturally occurring pigments and are mainly divided as betacyanins and betaxanthins. These pigments impart red-purple and yellow-orange colors to fruits and vegetables, respectively [64]. These compounds are commonly found as a pigment in *Beta vulgaris* (beetroot), in some fruits like *Opuntia* (prickly pears), *Hylocereus* and *Mamillaria* species [65]. Betalains are immonium conjugates of betalamic acid with cyclo-dopa and amino groups (amino acids, amines or their derivatives) [61]. The betalamic acid moieties conjugated with amino acids or amines, are termed betaxanthins. Condensation reactions between the betalamic acid and cyclo-dopa producing betanidin is a precursor of the betacyanins. Due to the presence of glycosylation or acylation, betanidins exhibit 29 different structures in nature. Betanins have also undergone several chemical reactions (isomerization, deglycosylation, hydrolysis, decarboxylation, and dehydrogenation) [66], but no reported study specifies the number of isomers or degradation products of betanins.

Several studies have been carried out on the extraction of betalains from vegetal wastes [25–33]. Kushwaha et al. [67] optimized an ecofriendly method for the extraction of betalains from beetroot pomace by changing different experimental variables such as the solid to liquid ratio, time, temperature and pH. Results showed the yield of betacyanin and betaxanthin to range from 1.75–62 and 1.79–61.62 mg/L, respectively. Similarly, the water extract of red dragon fruit (*Hylocereus polyrhizus*) peels were observed to have a betalain content of 30.18 mg/100 g of dry peel [26]. Further, microwave assisted extraction of betalains (9 mg/L of betalain content) from dried peel powder of dragon fruits has also been optimized [68]. Melgar et al. [69] extracted betalains from peels of *Opuntia engelmannii* using ultrasound and microwave-assisted extraction and reported betalain content to be 201.6 and 132.9 mg/g, with and without methanol, respectively.

Carotenoids

Among natural pigments, carotenoids are the most vital phytochemicals which are lipophilic in nature accounting for the red, yellow or orange color range in a variety of fruits and vegetables. Carotenoids widely occur as all-trans and cis-isomers forms [70]. Carotenoids are mainly categorized into two: carotenes (α-carotene, β-carotene and lycopene); and xanthophylls (lutein, zeaxanthin, and β-cryptoxanthin). Both carotenes and xanthophylls contain chains of hydrogen and carbon, but the presence of hydroxyl groups which represent oxygenated carotenoids in structure, to differentiate them [71]. Paprika is one of the rich sources of red carotenoids [50]. The capsanthin pigment is found in the fruits of *Capsicum annuum* (paprika) or *Capsicum frutescens* (piri piri). As per EU food legislation, capsanthin is listed as a natural food colorant (E160c), however, in USA legislation, oleoresin of paprika is permitted as a food colorant [72]. Commercially available carotenoids used as food coloring compounds are usually synthesized chemically [73]. However, currently, to boost green consumerism, these pigments can be produced from wastes generated by the fruit and vegetable processing industry. For example, paprika waste, (lutein-232.60 μg/g) [50], tomato peel (carotenoids-253.5 μg/g) [47], carrot peel [42] and their by-products (carotenoids-82.66 μg/g) [74]. Further, in spinach by-products, lutein recovery is reported to be 70% and chlorophyll recovery 96% [48]). Vegetal wastes are rich sources of natural carotenes exhibiting provitamin-A activity. In addition, seeds of *Bixa Orellana* (achiote), a source of annatto (natural pigment), has been used to color dairy formulations. However, there are no reports available on the natural pigments obtained from wastes or by-products of this plant, which should be explored in the near future.

Several studies have been reported on the extraction of carotenoids from vegetal wastes [34–39]. Recently, Tiwari et al. [74] extracted carotenoids from carrot pomace using a green extraction approach (flaxseed oil as green solvent) in combination with innovative extractions (ultrasonication, high shear dispersion) techniques. They concluded that the biorefinery approach is helpful to improve the extraction of carotenoids (82.66 μg/g) and β-carotene (78.37 μg/g) from carrot pomace. Goula et al. [39] adopted green extraction methods (sunflower oil and soy oil as green solvents) in combination with ultrasound for the extraction of carotenoids from pomegranate peels (0.6134 and 0.6715 mg carotenoids/100 g of dry peels in sunflower oil and soy oil, respectively). Kang et al. [50] optimized the method for lutein extraction by accelerated solvent extraction using response surface methodology from paprika leaves waste. These researchers reported the lutein content of paprika leaves extracted in ethanol to be 232.60 μg/g of the leaves. Kehili et al. [45] extracted lycopene and β-carotene from tomato by-products using supercritical CO_2 extraction and reported the recovery range to be 32 to 61% for lycopene and 28.38 to 58.8% for β-carotene. Similarly, Wang [46] extracted β-carotene (18.50 mg/100 g sample) from pumpkin seeds using supercritical CO_2 extraction. On the other note, de Andrade et al. [47] considered on carotenoids recovery (by supercritical fluid extraction) from various vegetal wastes. They reported on waste from the peel of the sweet potato, tomato, apricot, pumpkin and peach peels and pepper wastes (165.1, 253.5, 285.1, 142.0, 59.5, and 109.2 μg/g dry weight basis, respectively). Recently, ionic liquids in combination with ultrasonic-assisted extraction have been introduced for the extraction of carotenoids from the orange peel [75].

Chlorophyll

Chlorophylls is an oil-soluble, amphiphilic green pigment which is extensively dispersed in plants [76]. Chlorophyll molecules constitute a hydrophilic (porphyrin) group head and a lipophilic hydrocarbon tail (phytol group). Similar to lutein, due to its lipophilic hydrocarbon chains as phytol tail, it is generally considered insoluble in polar solvents [77]. Chlorophyll, found in plant foods are of two types: chlorophyll a and chlorophyll b. At position 7-carbon, methyl group (-CH_3) it is present in chlorophyll a whereas an aldehyde group (-CHO) is present in chlorophyll b. The difference in the structure leads to different colors of chlorophylls: blue-green (chlorophyll a) or yellow-green (chlorophyll b) [78]. Chlorophyll is well recognized, natural, green permitted food colorant (EU standard number is E140) [79]. According to the US legislation for natural food colorants published in Title 21 CFR 73, only copper chlorophyllin (water soluble) is authorized as a natural green food-colorant (CFR Section 73.125) [80]. Demand for chlorophyll is continuously increasing, corresponding to enhanced awareness on the use of natural colorants as well as health benefits imparted by them.

Chlorophylls are used for coloring food formulations and can be extracted from many types of vegetal wastes. However, there are no reports on the utilization of vegetal wastes for the extraction of chlorophylls and their further application as a colorant in food formulations. Limited available studies have explained the coloring attributes of chlorophyll. Considering the extraction of chlorophyll as an antioxidant, Zeyada et al. [43] have analyzed methanolic extracts of cucumber and watermelon peels for their chlorophyll content using HPLC and have recorded the content to be 3.46 and 5.28 mg/g (d.w.) respectively. They reported that except for the tomato peel, cucumber and watermelon peels exhibit the highest antioxidant activity among all the tested vegetable varieties, which was attributed to a high chlorophyll content. According to the study of Wang et al. [81] results obtained from eight varieties of avocado (*Persea americana*) peel and indicated them to be a good source of green pigment (Chlorophyll) (range of 25 to 66 μg/g) . Hence, avocado, being a rich source of green chlorophyll pigment, can be a platform to fortify or use as a coloring agent in various food formulations. Derrien et al. [48,49] optimized the extraction methods for lutein and chlorophyll from spinach (*Spinacia oleracea*) by-products with supercritical CO_2 [48] and green solvent extraction [49] using response surface methodology. Under optimum conditions, they reported the extraction yield for lutein and chlorophyll to be 72% lutein and 50% of chlorophyll [48], whereas, 70% for lutein and 96% for chlorophyll were recorded using green solvent extraction [49].

According to these authors' knowledge, there are no studies available which is practically based on the valorization of vegetal wastes for chlorophyll extraction. Hence, this review discusses avenues for future exploitation of the vegetal wastes in order to obtain natural green pigments. The addition of these green pigments valorized from vegetal wastes, in food formulations, synergizes the coloring attributes with an antioxidant potential which mainly favors the green consumerism and limits the application of synthetic coloring compounds in food.

Extraction techniques

Extraction of pigments involves the production of natural pigments for food applications. For the extraction of natural colorants, the first step is to obtain the crude pigment from the plant resources. Usually, conventional methods such as Soxhlet extraction techniques using organic/inorganic solvents, maceration or hydro-distillation have been widely used for natural pigment extraction [82]. For conventional extraction, generally water or diluted alcohol are employed for water-soluble pigments, while non-polar solvents are used for the extraction of lipophilic pigments [83]. The non-polar solvents used for the extraction of pigments are mainly of petrochemical origin e.g. hexane (carotenoids from Gac fruit peel [37]), acetone (lycopene from tomato pulp waste [84]), methanol (anthocyanins from eggplant peel [5]), trifluroacetic acid (betalains from pitaya fruits peel [85]). Most of them are mainly toxic in nature, though owing to their volatile nature, they can efficiently dissolve target pigments to facilitate convenient removal. Rather than being technically advantageous, traces of these solvents are unsafe for human consumption and will lead to carcinogenic risks in humans and can also contribute to environmental pollution [82].

Concurrently, traditional or conventional extraction methods have been escorted with assisted extractions techniques impregnated with other physical methods. Moreover, conventional extraction techniques that have shown a low extraction proficiency, are complex, and require a longer extraction time and huge capital investments. With these drawbacks, environmentally friendly innovative techniques are needed to allow safe extraction by using green solvents obtained from natural resources like plant based edible oils, wood etc. The use of greener extraction techniques is now considered to be an emerging, re-discovered and innovative method gaining much importance aimed toward avoiding the adverse effects of synthetic solvents. Nevertheless, these green extraction modes can also improve the extraction efficiency of natural pigments from vegetal sources.

Green extraction techniques

Many studies have been undertaken on the extraction of natural pigments by using synthetic solvents, but in this section, the focus is mainly on green technologies. Table 2 summarizes the greener extraction techniques that assist with various innovative techniques such as: ultra-sound-assisted extraction (UAE), microwave-assisted extraction (MAE), enzyme-assisted extraction (EAE), supercritical fluid extraction (SFE), pressurized liquid extraction (PLE), and pulse-electric field-assisted extraction (PEF) employed for the extraction of natural pigments from vegetal wastes and by-products [86–96]. Greener extraction techniques involve solvents such as: ionic liquids (ILs), water, ethanol, esters of fatty acid or oils of fruits and vegetables (soybean, rapeseed oil, cocoa oils etc.), glycerol etc. which are all gaining importance for the extraction methods for natural pigments. Many researchers are employing green solvents and other organic methods for the extraction of natural pigments [97]. Non-polar solvents used for the green extractions of lipophilic pigments having GRAS status incuding cyclopentyl methyl ether (CPME), 2-methyl tetrahydrofuran (MeTHF), dimethyl carbonate (DMC), ethyl lactate, ethyl acetate, α-pinene, D-limonene and plant based essential oils [98]. One more opportunity can be the use of a multi-stage extraction techniques which can be timely assisted by different physico-chemical approaches which may selectively target the desired pigments.

In Figure 2 some of the innovative extraction techniques/strategies *viz.,* UAE, MAE, SFE, PLE, PEF, and ILs which are proposed as substitutes to the conventional methods is depicted. These techniques are in demand due to their several advantages like the requirement for minimal solvents, are fast, convenient, can increase the extraction yield, protect pigments from degradation, enhance the quality of natural colorants, and are eco-friendly than conventional extraction techniques [99].

The toxicity of solvents used during extraction has encouraged the advancement of greener techniques

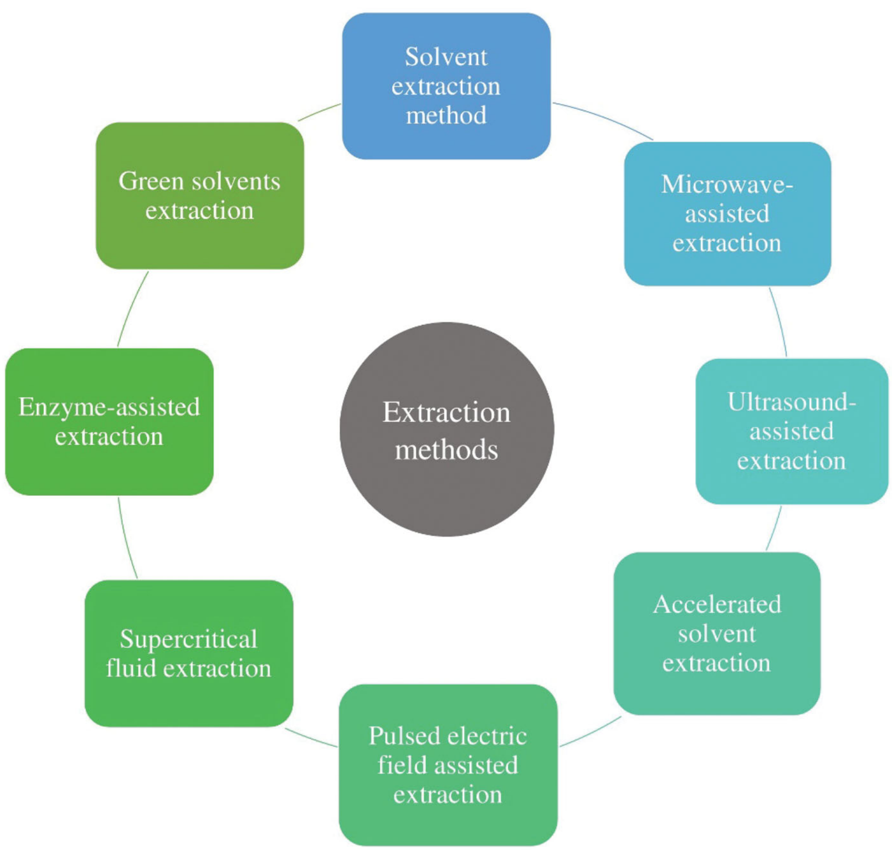


Figure 2. Different extraction methods recommended/commonly employed for extraction of natural pigments from vegetal wastes.

Table 2. Various green extraction techniques to extract pigments from vegetal waste.

Technique	Concept	Vegetal Waste Source	Pigment Extracted	Yield (Green Extraction)	Yield (Conventional Extraction)	Advantages	Drawbacks	Reference
Ionic Liquids (ILs) [BMIM][PF6], [BMIM][Cl], [HMIM][Cl], [BMIM][BF_4]	Usage of ILs to replace traditional solvents in order to selectively dissolve and extract bioactive compounds	Tomato wastes Orange peel	Lycopene Carotenoids	5.56 µg/g 32.08 µg/g	3.65 µg/g (in acetone) 7.88 µg/g (in acetone)	Chemically inert, thermally stable, high extraction rates environmentally safe,	High preparation cost (nor economical)	[86] [75]
Supercritical fluid extraction (SFE)	Extraction by varying pressure and temperature while using a medium such as CO_2.	Tomato peels	Lycopene	728.98 µg/g	608 µg/g (in hexane) 284 µg/g (in ethanol) 320 µg/g (in ethyl acetate)	Environmentally safe, fast, highly selective	Greatly impacted by the property of the fluid used	[87, 45],
Pressurized liquid extraction (PLE) or Accelerated solvent extraction (ASE)	Using high pressure to extract the compounds, while using higher temperatures	Blackberry residues	Anthocyanins	6.33 mg/g	5.02 mg/g	Faster extraction, reduced solvent usage, higher operating temperature increases diffusion rate	Extraction efficiency greatly varies with temperature, pressure and type of solvent used	[88]
Ultrasound assisted extraction (UAE)	Leaching of bioactives from plants *via* cavitation facilitated by using ultrasound sounds (20 kHz to 100 MHz).	Tomato wastes	Lycopene	5.11 mg/g	1.23 mg/g	Analytically simpler, more efficient, lower extraction temperature, faster processing time	Higher cost, specialized setup required	[89–90]
Microwave assisted extraction (MAE)	Penetration of solvent into waste solid matrix assisted by microwaves (300 MHz to 300 GHz)	Black currant marc	Anthocyanins	20.4 mg/g	12.5 mg/g	Maximum anthocyanins yield obtained after 10 min using MAE, while using conventional extraction after 300 min, lesser solvent quantity used in MAE	Energy consuming, External factors (temp., pressure, power freq.) greatly affect the output and efficiency	[91–92]
High voltage electric discharge	Extraction of compound by damaging the cell structure to form gaseous cavities followed by phase decomposition	Grape stem extract	Polyphenols	6.6 g/100g	4.9 g/100g	Efficient, enhanced mass transfer, lower operating temperature, shorter extraction time	Voltage fluctuations greatly affect yield, Can be expensive	[93–94]
Pulsed Electric Field (PEF)	Applying a pulse of electric field (100-300 V/cm to 20-80 kV/cm) on the feed material put between two electrodes at room temperature	Beetroot Grape by-product	Betalains Anthocyanin	90% (300 min) 14.05 mg/g	20% (300 min) 7.93 mg/g	Enhanced extraction from plants, reduce time of extraction	High energy consumption	[95] [17]
High hydrostatic pressure (HHP)	Application of relatively higher pressure (100-1000MPa) at 0 °C-100 °C for a short duration.	Grape by-products	Anthocyanins	11.21 mg/g	7.93 mg/g	Enhanced mass transfer, energy efficient, better secondary metabolite diffusion	High initial setup cost	[17, 96]

and materials to extract bioactive compounds from vegetal wastes and by-products. Moreover, termed as cold extraction methodologies, these methods maintain the natural composition and stability of extracted compounds while requiring lower energy, less number of process steps, a reduction in setup size, better mass and heat transfer and higher yield [100–101].

Yamashita et al. [10] developed an easy and cost-effective method by using less toxic extractants for the extraction of anthocyanins from blackberry by-products. Frozen blackberry pulp by-products were extracted with water (1:3 ratio, respectively) for anthocyanins, and concentrated in a rotatory evaporator at 60 °C. The aqueous and concentrated extracts had a higher anthocyanin content (718 and 389 mg cyanindin3-glucoside/100g of d.w.). Derrien et al. [49], utilized green extraction technique assisted with supercritical CO_2 (SC-CO_2) extraction using 93% ethanol for the extraction of chlorophyll and lutein from spinach by-products/wastes and confirmed higher recovery of phytopigments (70% for lutein and 96% for chlorophylls) when compared with conventional extraction using acetone.

Parra-Campos & Ordóñez-Santos [12], extracted anthocyanins from coffee exocarp and observed the highest content of anthocyanins (0.145 mg cyd 3-glucoside/g) obtained with 60% ethanol used as an extracting medium and with a 25% solid solvent ratio. Monrad et al. [11] contemplated the extraction of anthocyanins from red grape pomace by utilizing expeller techniques. These authors have recorded an increase in the yield of anthocyanins (68 and 41% for crude and dried sample, respectively). In a study by Drosou et al. [24] three different extraction methods (Soxhlet, MAE and UAE) along with solvent extraction (ethanol, water and their mixture-1:1), were employed to extract anthocyanins from the pomace of 'Agiorgitico' red grapes. Results of this study indicated highest procyanidins (43469 ppm) and anthocyanin contents (34188 ppm) in UAE- assisted techniques with solvents in combination (water: ethanol) compared to all treatments. Vulić et al. [31] extracted betalains from the peel and pomace of beetroots by using a mixture of water and ethanol (1:1) acidified with acetic acid (0.5%) and assisted with ultrasonic extraction (50–60 Hz, 22 °C, 125 W, 30 min). They reported three major betalains from beetroot peel waste: betanin (3.8 to 7.5 mg/g), isobetanin (1.2 to 3.1 mg/g) and vulgaxanthin (1.4 to 4.3 mg/g). Beetroot pomace extract showed significant amounts of betanin (37.22 mg/100g) (all on dry weight basis). Recently, Goula et al. [39] have developed a new process for the extraction of carotenoids from pomegranate peel waste by utilizing UAE assisted extraction with vegetable oils (sunflower and soybean oils) as a green solvent. They reported pomegranate peels to be an enriched source of carotenoids (0.6134 and 0.6715 mg carotenoids/100 g of dry peels with sunflower oil and soy oil, respectively). Working with tomato wastes, Strati et al. [102] utilized pressurized liquid extraction (PLE) assisted with ethanol at a pressure about 700 MPa/10 min, and reported a higher content of total carotenoids (9.30 mg/kg d.w.) and lycopene (7.04 mg/kg d.w.) as compared to conventional extraction (total carotenoids- 3.63 mg/kg dw, lycopene- 2.47 mg/kg dw). Corrales et al. [17] compared a pulsed electric field (PEF) assisted extraction with two other extraction techniques (ultrasonics and high hydrostatic pressure, HHP) for anthocyanins extraction from grape by-products. They observed an improvement in extraction efficiency (10% and 17%) in PEF technique compared to HHP and conventional solvent extraction. Results showed increased antioxidant activity; about four-fold in PEF, three-fold in HHP and two-fold in with ultrasonic treated samples when compared to conventional solvent extraction techniques (70 °C for 1 h). In another study, Kehili et al. [45] explored SC-CO_2 techniques assisted with solvent extraction techniques and using three solvents (hexane, ethanol and ethyl acetate), for the extraction of lycopene and β-carotene from tomato peel. Extraction with SC-CO_2 yielded lycopene about 728.98 mg/kg dry basis under suitable processing conditions (pressure- 400 bar, CO_2− 4 g/min at 80 °C). In contrast, extraction with solvents (hexane, ethanol and ethyl acetate) were recorded to have a lower lycopene content (608, 284 and 320 mg/kg dry basis, respectively). Enzyme-assisted extraction (cellulase and pectinase), assisted with ethyl lactate, resulted in an increase in the yield of carotenoids (six-fold) and lycopene (ten-fold) was reported for tomato waste when compared with non-enzyme treated samples [102]. This study suggested that the use of commercially cheaper food-grade enzymes have huge potential to be explored on a pilot scale for the extraction phytopigments from agri-food wastes and by-products.

Recently, ionic liquids (ILs) have been used on various raw materials to obtain bioactive compounds, carotenoids being one such example [103]. Martins and Rosso [86], reported the use of IL [BMIM][PF_6] (1-butyl-3-methylimidazolium hexafluorophosphate) and [BMIM][Cl] (1-butyl-3-methylimidazolium chloride) to extract lycopene from tomato waste and reported higher amounts (5.56 μg/g) of lycopene as compared to acetone (3.65 μg/g). Murador et al. [75], demonstrated a novel method to extract carotenoid from orange peel, using four different ILs ([BMIM][PF6], [BMIM][Cl], 1-hexyl-3-methylimidazolium chloride ([HMIM][Cl]) and

1-n-butyl-3-methylimidazolium tetrafluoroborate ([BMIM][BF_4])), assisted by ultrasound. [BMIM][Cl] was determined to be the most effective at extracting the carotenoid (32.08 μg/g), while acetone could only extract 7.88 μg/g of dry matter.

Some of the available innovative techniques along with organic solvents again cause an ill-impact on health even though in minimal quantity. Studies are ongoing with continuous efforts made to search new economical methods suitable for the extraction of all the pigments with improved quality following extraction. More research activities need to be undertaken aimed toward avoiding the use of synthetic colorants in foods to reduce the impact of synthetic colorants on the human body.

Considerable research works is lacking in the area of green technologies assisted with innovative technologies for the extraction of natural pigments on an industrial scale. There is a need to analyze the extraction costs for different phytopigments extracted from various green extraction methods and to identify the most feasible technique which can be best suitable for all the applications on an industrial scale.

Encapsulation of natural pigments

Stability is one of the vital aspects to be considered for the utilization of natural pigments as colorants and antioxidants in food formulations. The strength and stability of natural pigments are affected by several factors during processing and storage. Natural pigments extracted from plant resources or agri-food wastes are highly unstable and are susceptible to degradation by external (processing conditions such as temperature and pH) as well as internal factors (e.g. concentration of pigment). The naturally occurring pigments are inclined to degrade easily in aqueous and lipophilic solutions [104]. The stability of these pigments can be increased by various concentrations in the system; low pH and temperature; presence of stabilizers (e.g. chelating agents, antioxidants); absence of light and the presence of acylation or glycosylation in the structure. In this way, micro or nanoencapsulation represents a promising concept and a best-suited technique for the entrapment of natural pigments in a coating enclosure to enhance their shelf-life [105]. This is a technique which entraps the natural pigments using biopolymers to protect them from various processing and environmental hazards such as: moisture, oxygen, temperature, light, etc. This further improves their stability and ensures easier handling by changing them from liquid to powder forms [106]. The potential of encapsulation techniques aimed to preserve the natural pigments extracted from vegetal wastes is are summarized in Table 3 [107–115]. Several studies have reported on the use of micro/nanoencapsulation techniques such as spray drying [108–114], freeze-drying [6,10,14,19,21,23,108,109,112,113], electrospinning-nanofibers [34] or complex coacervation- microsphere [52] for the encapsulation of natural pigments. Spray drying is the best technique for the encapsulation of natural colorants because of it is a minimal exertion, rapid, and a reliable modern technique. During spray drying, the free-flowing ability, quality, powder efficiency, and bio-activity of natural pigments after drying treatment depends on the processing conditions such as temperature, atomization speed, feed rate, and carrier to feed ratio [116]. Microencapsulation represents a promising application to the modern-day food industry by encapsulating unstable natural pigments which can help in enhancing the stability while maintaining their color during processing conditions. Microencapsulated powders obtained from natural pigments extracted from wastes or by-products can be utilized in the formulation of several types of functional foods and beverages such as: confectionery, chocolates, jellies, sauces, ice-creams, candies, juices and instant drink powders to color the food commodities and enrich the health-promoting potential.

Bioactivities and health benefits

Food industries are using pigments as color intensifiers, additives as well as antioxidants. There is a need to explore various vegetal waste resources to obtain food-grade pigments with bioactive potentials. This is mainly due to consumers' awareness on health and numerous benefits delivered by these compounds (see Table 1, Figure 3). These pigments possess various biological roles in the protection of human health, like anti-inflammatory (utilizing cytokines signaling) [117], antioxidant (utilizing free radical scavenging pathway) [118], anticancer properties, antimicrobial, cardioprotective and antithrombotic (utilizing mitogen-activated protein kinase pathway), improving ocular and neural health and the prevention of non-communicable diseases (utilizing cyclo-oxygenase pathway) [25,119].

Antioxidant activity

The majority of the wastes and by-products have been reported to be a rich source of bioactive antioxidant compounds. These antioxidant rich compounds have been mainly isolated from berries (e.g. ribes,

Table 3. Various techniques recommended to encapsulate natural pigments from vegetal wastes.

Encapsulates/Extract	Target Pigment	Source	Concentration added	Coating materials	Food applications	Outcome	Reference
Freeze drying	Anthocyanins	Blackberry by-products	–	Maltodextrin- 10DE and 20DE	–	Microcapsules of MD-10DE darker, than MD-20DE Difference in DE affects the tonality	[10]
Spray-drying, Freeze-drying, Supercritical antisolvent	Anthocyanins	Blackberry residue	–	Polyvinylpyrrolidone (PVP)	–	No degradation of anthocyanins, PVP increases the thermal stability of anthocyanins	[6]
Freeze-drying	Anthocyanins-rich extract	Jaboticaba pomace	–	Maltodextrin, pectin and soy protein isolate	–	Carrier protect the anthocyanins under UV treatment	[19]
Rapid Extraction of Supercritical Solution (RESS) & Ionic gelation	Anthocyanins	Jaboticaba peel	–	Polyethylene glycol, ethanol as co-solvent	–	Color retained at T: 323.15 K; P:10MPa	[20]
Spray drying	Anthocyanins extract	Blueberry pomace	–	Whey proteins	–	Anthocyanins degradation followed the first-order kinetics, antioxidant capacity increased	[107]
Spray and freeze drying	Anthocyanins content	Acerola residue- extract	–	Gum Arabic and Maltodextrin	–	Acerola residue after microencapsulation retain its antioxidant activity	[21]
Spray and freeze drying	Anthocyanins extract	Blueberry pomace	–	Protein sources: wheat flour, chickpea flour, coconut flour, soy protein isolate	–	Spray drying protected anthocyanins from degradation during 16 weeks at 4 °C and 20 °C	[108]
Spray and freeze drying	Anthocyanin extract	Bordo grape skin	–	Gum arabic, partially hydrolyzed guar gum, and polydextrose	–	Retention of anthocyanins was higher (99.58–80.75%), for spray-dried treatment made with PHGG	[109]
Extract as pigment	Anthocyanins	Coffee exocarp	3% extract	–	French meringue	Acceptable color as control samples	[12]
Spray drying	Anthocyanins extract	Eggplant peel	0.5-2% peel powder	Maltodextrin and Gum Arabic	Gummy candy	0.5 and 1% peel powder-candy with acceptable color and taste	[13]
Freeze-dried	Anthocyanins - extract	Sour cherry pomace	10% and 15% encapsulated powder	Whey and soy proteins	Cookies	Satisfactory sensory acceptance, Increase shelf-life	[14]
Freeze-drying	Anthocyanins - extract	Sweet cherry skins	5% and 10% encapsulated powder	Whey proteins isolate and chitosan	Yoghurt and marshmallow	Anthocyanins decreased in yoghurt and increased in marshmallows during storage	[23]
Microencapsulation-spray drying	Betalain Extract	Jaboticaba peel	2% and 4%	Maltodextrin	Fresh pork sausages	Reduce the lipid oxidation of the sausages during 15 days of storage at 1 °C	[110]
Microencapsulation-spray drying	Betalains	Pitaya peel extract	0–1000ppm	Maltodextrin	Pork patties	Reduced the hardness and chewiness changes in the pork matrix	[33]
Spray drier	Carotenoid-rich oil	Gac fruit peel	–	Whey protein concentrate and gum Arabic	–	Higher retention of antioxidant capacity and carotenoids content after encapsulation at storage temperatures.	[111]

(*continued*)

Table 3. Continued.

Encapsulates/Extract	Target Pigment	Source	Concentration added	Coating materials	Food applications	Outcome	Reference
Spray and freeze drying	Carotenoids content	Acerola residue- extract	–	Gum Arabic and Maltodextrin	–	Acerola residues after microencapsulation retain its antioxidant activity	[21]
Spray and freeze drying	Carotenoids	Red pepper waste	–	Whey protein	–	Rapid initial release of carotenoids from whey protein matrices during bioavailability studies	[112]
Spray and freeze drying	Carotenoids	Red pepper waste	10% (w/v)	whey proteins isolate and sunflower oil	Yogurt	Retained carotenoids with improved nutritional, color and bioactive properties	[113]
Spray drying	β-carotene	Peach palm residues (*Bactris gasipaes*)	–	Gum arabic, gelatin, sugar and lecithin	–	Better β-carotene stability with higher levels of retention (88.24%),	[114]
Electrospinning-nanofibers	Lycopene	Tomato peel	–	Gelatin	–	Encapsulated-gelatin fibers stored *at* – 20 °C showed highest retention of lycopene	[34]
Extract	Lycopene	Tomato pomace	50-200 mg/ kg	–	Margarin, cream, cheese spread, ice cream, sponge cake, Kunafa dessert, meat sausage, cooked rice, popcorn, extruded snack food and fruit juice	No off flavor, no objectionable color	[115]
Extract	Lycopene	Tomato peels	1–5 % extract	–	Ice-cream	Addition of 3 and 2% of extract had the highest scores for sensory analysis	[36]
Complex coacervation-microsphere	Lycopene	Tomato skin	2 g/L microspheres	TiO_2 nanoparticles coacervated with gelatin–pectin complex	Dipping Fresh cut apple slices	Reduced apple browning after 9 days (5 °C), Improved bioactive quality of lycopene during shelf life studies on fresh cut apples (20 mg/kg lycopene after 9 days).	[52]

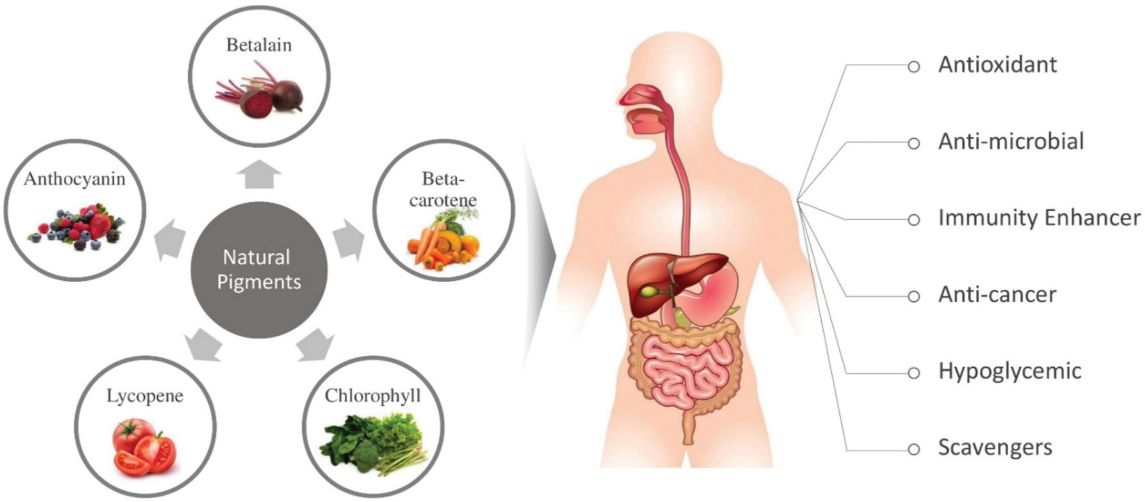


Figure 3. Natural pigments from vegetal wastes and their proved bioactivities.

chokeberries, blueberries, blackberries and raspberries). Nutritional properties of these waste materials have turned them into promising sources of functional compounds. Usually, the waste discarded from fruits and vegetables contains high amounts of antioxidant compounds [120]. Anthocyanins, flavonoids, vitamin E, C, phenolic compounds, fibers, carotenoids, and other antioxidants are the key bioactive compounds present in these waste materials, thus making them valuable sources of these compounds. These compounds can thus be extracted to be reused as functional food ingredients in other food products and supplements to provide much required antioxidants and to provide health benefits.

Seed pomace waste of both raspberry and blackberry can be a good source antioxidants. Besides containing omega-3 (α-linolenic) and omega-6 (linoleic) (ratio 1: 2–4), these oils can be a potential source of bioactive compounds such as: phenols, tocopherols, carotenoids and sterols, which are recognized antioxidant agents. The presence of these antioxidants in the oil extracted from raspberry and blackberry seed pomace have elevated the usage of these seemingly waste products [121]. Many health-promoting compounds can also be extracted from leaves as well. Cranberry leaves and pomace have been shown to contain more polyphenols resulting in the elevated antioxidant properties when compared with fruits, thus making them an alternative source to extract these beneficial compounds [122]. Orange peel has a lower antioxidant potential than flesh wastes. Though both extracts have been shown to be effective in protecting against hydrogen peroxide free radicals, induced DNA damage on human leukocytes [123]. Further, in a recent study on pomegranate extract, the presence of ellagic acid is reported to show antioxidant properties [124]. It has also been reported that the presence of natural antioxidant additives in the wastes of tomato and potato renders them to be an excellent protective agent aimed to preserve vegetable oils (by reducing the oxidation of oils) [125].

Anti-cancer activity

Natural plant based pigments have been well established to be a potential anti-cancer agent. Núñez Selles et al. [126] have explored the application of mangiferin – a naturally occurring bioactive xanthonoid extracted from the mango tree for cancer treatment. Besides being a good antioxidant and anti-inflammatory agent, *Mangiferin,* when used either individually or in conjunction with other anti-cancer chemicals, is potentially beneficial in treating lung, brain, cervix, prostate and breast cancers, and leukemia. On the other note, berry extracts have been proven to exhibit anti-tumour properties due to the presence of polyphenols, ellagitannins and anthocyanins [127]. Black raspberry seed wastes can cause apoptosis in colon cancer cells (HT-29) resulting in suppressed cellular spread [128]. In freeze dried apple wastes, the inhibitory effects of non-extractable antioxidants was more pronounced than the extractable antioxidants on human cancer cells (viz. HepG2, HeLa, and HT-29) [129]. It has been proved that ellagitannins extracted from pomegranate are useful in prostate LNCaP and breast MCF-7 cancer cell growth [130]. Production waste of tomato juice exhibited anti-cancer properties *via* superoxide and hydroxyl anion radicals, which inhibited tumorous cell growth of MCF7, HeLa, and MRC-5 [131]. Betalains extracted from beetroot pomace have been shown to be responsible for their potent antioxidative and anticancer properties, which exhibited significant proliferative effects in human cell lines- MCF7 and MRC-5 [132].

Anti-inflammatory

Various phytochemicals including those of natural pigments from fruits and vegetables have gained the attention as therapeutic agents, which has been proved to reduce inflammation. Besides, they are recommended to be used as analgesics and anti-inflammatory agents in food and pharmaceutical preparations. Pectinolytic preparations were used to extract polyphenolic compounds and anthocyanins to study the potential anti-inflammatory activities of enzymatically treated raspberry pomace [133]. These researchers found that anthocyanins thus obtained were able to inhibit lipoxygenase and cyclooxygenase 2 activities, resulting in the anti-inflammatory properties. Multiple studies have conclusively found that citrus pectin can regulate inflammatory response by affecting immune cells. *In-vitro* and *in-vivo* studies have found citrus pectin to induce endotoxin, causing alleviation of inflammatory sensations [134]. Another phytochemical, limonin (a triterpenoid) has been extracted from citrus waste [134]. Limonin can act as an anti-inflammatory, anti-bacterial, antioxidant and anti-cancer agent [135]. Huynh et al. [136] found cauliflower by-products, when subjected to solid state fermentation (using filamentous fungi) to produce significant amounts of phenolic compounds which increased the extractability of kaempferol glucosidase Kaempferol is a natural flavonoid that exhibits anti-carcinogenic and anti-inflammatory effects [137]. A key usage of peel extracts is its role as an inhibitor toward nitric oxide (an inflammation mediator) and cytokines TNF-α, which could be explained by a decrease in free radicals during inflammation [138].

Anti-microbial activity

Antimicrobial activity of fruit extracts (mainly of peel) is likely to be caused by the presence of colored pigments (anthocyanins, carotenoids, etc) which exhibits multiple antimicrobial mechanisms and synergies inside a microbial cell. Avocado peel extracts exhibited elevated antimicrobial potential than nisin, a naturally occurring dipeptide with antimicrobial properties. Additionally, avocado peel extracts also exhibited antimicrobial properties against *Listeria innocua (L. innocua), Escherichia coli (E. coli)*, *Leuconostoc mesenteroides, Weissella viridescens* and *Lactobacillus sakei*. When it is combined with 39% nisin peel extracts it displayed the highest antimicrobial impact on *L. innocua* [139]. In general, the antibacterial impact of peel extract was not only significant against Gram-positive bacteria, but also on *E. coli* [140]. Kosińska et al. [141] described that the polyphenol and tannin content of avocado seed extract had an antimicrobial effect on *Staphylococcus epidermidis, Listeria monocytogenes* and *Mycobacterium avium*, thus concluding that urinary tract infections could be prevented by the tannins present in avocado seed extracts [141]. Kapadia et al. [142] brought forward the antimicrobial effects of banana peels extracts on *Aggregatibacter actinomycetemcomitans* and *Porphyromonas gingivalis*. By-products from grape processing have been used to extract products that can serve as antimicrobial agents and natural antioxidants in fruit juices and meat products [143]. For instance, grape seed oil has been proven to have antimicrobial properties, as demonstrated by Garavaglia et al. [144], along with other health benefits. Beetroot pomace has been used in many functional supplements and foods due to its antimicrobial impact [67]. By-products of cauliflower were evaluated for their antimicrobial impact on *L. monocytogenes*. The results of this study demonstrated its antimicrobial effects rendering it to be an important source to create biobased preservatives for ready-to-eat refrigerated foods [145].

Anti-obesity activity

The natural phytopigments (mainly anthocyanins and carotenoids) are envisaged to play a significant role in lessening obesity contributing toward a decrease in adipose tissue (fat deposition). Various components and extracts from mango wastes have exhibited anti-oxidative and anti-inflammatory properties and at the same time have shown promising results at reducing obesity and diabetes in humans and animals. They have also proven to be good at: neuroprotection, managing intestinal health, prevention of skin cancers and cardiovascular diseases (constructive impact on the microbiota of gut) [146]. Alam et al. [147] demonstrated the anti-obesity effects of hydroxycinnamic acid derived from the Mavolanate-Shikimate biosynthesis pathways in plants. Betanine extracted from beetroot waste can serve as an excellent alternative route to prevent cancer and other disorders such as: Alzheimer's disease, diabetes, and obesity [148]. Gao et al. [149] observed the impact of betanine intake on a large general population, and found direct correlation between betaine consumption and better body composition. Devalaraja et al. [150] reported persimmon (*Diospyros kaki* L.) fruit peel extract to contain bioactive proanthocyanidins which exhibited anti-diabetic and anti-obesity effects. Apple pomace was also found to be rich in Phlorizin and Phloretein which suppressed obesity [151].

Neuroprotective activity

The extracts rich in natural pigments have been proved to exhibit higher neuroprotective activity than polyphenols. The bioactivity of banana peel extract has been found to deliver strong neuroprotective effects, among other advantages, such as: antioxidant, antibacterial, antifungal and others [152]. D-Psicose is another product extracted from fruit and vegetable wastes that have been found to exhibit multiple physiological advantages such as lower glycemic levels, suppress blood glucose, neuroprotective impact and an anti-dyslipidemic [153]. Angeloni et al. [154] studied the bioactivity of phenols derived from olive oil, finding its advantageous effects in recovery from: ischemic brain injury, Alzheimer's, spinal cord injury and Parkinson's diseases.

Based on the above reported bioactivities, it is hereby opined that pigments obtained from vegetal wastes and by-products can exhibit rich bioactivities. Future research is warranted to explore the biological potential of these pigments to be utilized in food, pharmaceutical and cosmetic industrial applications.

Future prospects and conclusion

Vegetal wastes and/or by-products obtained from the food bioprocessing industry remains as a major leftover leading to environmental pollution. However, these wastes and by-products can be a valuable and promising source of natural pigments. New advances, innovations and challenges in the extraction and application of these natural pigments to harvest their potential in functional bioactive are the core focus of this review. This review offers certain safe, potential, biodegradable, pigment alternatives to the currently used artificial coloration paradigm, which are usually extracted from renewable waste resources and by-products from fruits and vegetable industry. Recent investigations on the extraction of natural pigments embrace the quest for more innovative and financially feasible resources, suitable extraction strategies, developments in bioprocessing technologies, related innovations in pigment stabilization, suitability of a method for a wide range of pigments, to obtain a better quality of colors, probability of developing pigment-enriched functional foods and bioactives. To meet the current demands, there is an urgent need for food industry to take steps to minimize the vegetal waste to exploit the natural pigments in a sustainable manner that are natural, cost-effective, biodegradable and those which facilitates the minimal production of harmful intermediates when they enter the ecosystem and/or the food chain.

There is still ample scope for research activities that can be undertaken to obtain a suitable, reliable alternative which is considered to be safe, highly efficient, non-solvent based, environmentally friendly model techniques for pigment extraction which can contribute for higher extraction yields. The extraction of natural pigments from vegetal wastes followed by the incorporation of food products not only improves the color of the food commodities but also enriches the antioxidant capacity of food, aiding the health benefits by chelating free radicals from the body when introduced in diets, improving an esthetic appeal to consumers, raising the monetary aspects for manufactures and the ultimate benefits to the ecological atmosphere by reducing the waste in the fruitful arena. Also, there is minimal information available on the utilization of pigment-encapsulated functional foods. Encapsulation of the natural pigments can enhance the stability of these compounds within the processing environment of a model food system. Efforts need to be made to develop greener technologies to extract natural pigments from vegetal wastes, micro/nanoencapsulation techniques to enrich the bioavailability and stability that can simultaneously provide health benefits. Thus, the selection of greener technologies, suitable encapsulating material, nature of the encapsulant, encapsulation methods, nature of the core (natural pigment) material, processing conditions suitable for natural pigments still remains a challengeable constraint.

Finally, it is concluded that exploitation of natural and health benefiting pigments from food wastes and by-products not only helps to minimize the environmental stress and supports the circular economy concepts, but can also helps the economic gains to the dependent industry.

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